(19) Europäisches Patentamt
European Patent Office
Office européen des brevets

(11) EP 0 692 148 B1

(12) # EUROPEAN PATENT SPECIFICATION

(45) Date of publication and mention of the grant of the patent:
01.07.1998 Bulletin 1998/27

(21) Application number: **94912182.6**

(22) Date of filing: **04.03.1994**

(51) Int Cl.[6]: **H01M 8/02**

(86) International application number:
PCT/US94/02347

(87) International publication number:
WO 94/23466 (13.10.1994 Gazette 1994/23)

(54) **LAMINATED ELECTROLYTE RESERVOIR PLATE**
BESCHICHTETE ELEKTROLYTBEHÄLTERPLATTE
PLAQUE STRATIFIEE DE RESERVOIR A ELECTROLYTE

(84) Designated Contracting States:
DE DK ES FR GB IT NL SE

(30) Priority: **31.03.1993 US 40955**

(43) Date of publication of application:
17.01.1996 Bulletin 1996/03

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- **PATENT ABSTRACTS OF JAPAN vol. 13, no. 147 (E-741) (3495) 11 April 1989 & JP,A,63 308 874 (SHOWA DENKO K.K.) 16 December 1988**

EP 0 692 148 B1

Description

Technical Field

The present invention relates to electrolyte reservoir plates, and especially relates to laminated electrolyte reservoir plates.

Background of the Invention

Fuel cells, particularly acid fuel cells, are comprised of an anode chamber, an anode electrode on a substrate, an electrolyte matrix, a cathode electrode on a substrate, a cathode chamber, an electrolyte reservoir plate, and a separator plate. These components are aligned electrically in series such that a stack of fuel cells can be employed in the production of electricity.

The electrolyte reservoir plate is a porous structure filled with electrolyte. During fuel cell operation, the electrolyte reservoir plate supplies electrolyte to the fuel cell to replenish electrolyte which has been lost by evaporation therefrom. Due to the constraints of the electrolyte reservoir plate formation process, these plates are costly to manufacture and possess limited strength.

For example, electrolyte reservoir plates can be formed in a dry-laid process where graphite powder, powdered phenolic resin, and fibers are showered onto a slow-moving belt to form a layer. The layer enters an oven where it is compacted with a second belt to form a 3.81 mm (0.150 inch) thick layer which is heated until the phenolic resin melts and coats the graphite powder and fibers. The resin is then cured, thereby bonding the graphite powder and fibers in a composite. Although this is a common electrolyte reservoir plate formation process, the forming speed is slow and it is difficult to incorporate relatively long fibers which are necessary for electrolyte reservoir plate structural integrity. Longer fibers tend to become entangled in the dry-laid feeder, thereby forming fiber bundles in the finished composite. This fiber bundling, which corresponds to uneven fiber distribution, creates weak areas within the composite which are susceptible to structural failure. Composite structural integrity is maximized at fiber lengths greater than about 1.0 mm (about 0.040 inches) while the dry-laid process is limited to fiber lengths of about 0.51 mm (about 0.02 inches).

An additional disadvantage of the dry-laid process is the post formation impregnation of two parallel edges of the composite with a substance such as hydrophilic ink to form a gaseous edge seal when filled with electrolyte. This prevents a possible mixing of fuel and oxidant utilized in the fuel cell. The impregnation, however, becomes increasingly difficult if the electrolyte reservoir plate thickness increases above about 2.54 mm (0.10 inches), if the density increases to about 1.0 g/cc or greater, if the median pore diameter decreases to below about 20 microns, and/or if any of these parameters are not substantially uniform throughout the composite. Consequently, the tolerances in the specification for the electrolyte reservoir plate are small and the fabrication is difficult, resulting in many rejected parts.

GB-A-2 177 387 and GB-A-2 177 388 disclose carbon-graphite components, in particular electrolyte reservoir layers, manufactured by carbonizing and graphitizing a precursor sheet of cellulose fibers, purified graphite particles and a carbonizable thermosetting resin. The documents are, however, silent about forming a laminated structure of precursor sheets, including main sheets and edge strips located at the perimeter thereof, and carbonizing and graphitizing the structure.

What is needed in the art is an improved electrolyte reservoir plate which is more efficient to process, and has improved structural integrity and gas edge barriers.

Disclosure of the Invention

The invention relates to a laminated electrolyte reservoir plate and a method for making the same. The electrolyte reservoir plate comprises a mixture of graphite powder, reinforcing fibers, cellulosic fibers, and thermosetting resin, which has been formed into a planar sheet. This planar sheet has been cut into a plurality of main sheets and a plurality of edge strips which have been laid-up, such that said edge strips are located along the perimeter of said main sheets along said opposing edges, laminated together, carbonized, and graphitized.

The method for forming the laminated electrolyte reservoir plate comprises mixing the graphite powder, reinforcing fibers, cellulosic fibers, and thermosetting resin with a liquid to form a slurry which is formed into a planar sheet. The liquid is then removed from the planar sheet and the sheet is cut into a plurality of main sheets and a plurality of edge strips. The main sheets and edge strips are laid-up, such that said edges strips are located along the perimeter of said main sheets adjacent to said opposing edges, laminated, carbonized, and graphitized to form the electrolyte reservoir plate.

An alternative method for forming the laminated electrolyte reservoir plate, comprises mixing the graphite powder, reinforcing fibers, and cellulosic fibers, with a liquid to form a slurry which is formed into a planar sheet. The liquid is removed from the planar sheet to form a dried sheet which is impregnated with thermosetting resin. The impregnated sheet is cut into a plurality of main sheets and a plurality of edge strips which are laid-up, such that said edges strips are located along the perimeter of said main sheets adjacent to said opposing edges, laminated, carbonized, and graphitized to form the electrolyte reservoir plate.

The foregoing and other features and advantages of the present invention will become more apparent from the following description and accompanying drawings.

Brief Description of the Drawings

Figure 1 is an exploded perspective view of the lay-up for the laminated electrolyte reservoir plate of the present invention.

Figure 2 is a frontal view of the lay-up shown in Figure 1.

The figures are meant to further illustrate the invention and not to limit the scope thereof.

Best Mode for Carrying Out the Invention

The present invention is directed toward a laminated electrolyte reservoir plate comprising graphite powder, cellulosic fibers, and reinforcing fibers all bonded together with a thermosetting resin. This electrolyte reservoir plate is formed in a papermaking process where the cellulosic fibers provide sufficient wet-strength in the fabrication process such that the paper, in the form of a wet, flat sheet, possesses sufficient tensile strength to traverse the entire paper-making machine without breaking. In contrast, the reinforcing fibers provide structural integrity to the finished electrolyte reservoir plate. The graphite powder provides increased thermal and electrical conductivity in the finished electrolyte reservoir plate as well as principally influencing the median pore diameter thereof, while the thermosetting resin binds the fibers and graphite powder together by forming a continuous phase throughout the electrolyte reservoir plate after lamination and provides an electrical and thermal bridge between the individual graphite particles for enhanced electrical and thermal conductivity after graphitization.

The graphite powder typically has a median particle size which compliments the median pore size of the adjacent electrode substrate based upon capillary forces. If the electrolyte reservoir plate possesses a median pore size smaller than the electrode substrate, the electrolyte reservoir plate will always retain the majority of the cell electrolyte in preference to the electrode substrate, while continuously replenishing electrolyte to the electrode substrate, edge seals, and electrolyte matrix which has evaporated from the cell. Therefore, since the electrode substrate has a median pore size of about 25μ to about 35μ, an electrolyte reservoir plate median pore size of about 10μ to about 20μ will attain optimum electrolyte sharing between the electrolyte reservoir plate and the adjacent electrode substrate. Consequently, the median particle size of the graphite powder is typically about 2 microns (μ) to about 300μ, with a median particle size of about 40μ to about 150μ preferred to attain a median pore size of about 10μ to about 20μ. These particle sizes are additionally chosen so that the electrolyte reservoir plate does not wick electrolyte from the matrix layer or edge seals of the cell which require nearly 100% electrolyte fill to prevent fuel and/or oxidant mixing.

The concentration of graphite powder in the final electrolyte reservoir plate is typically about 25 weight percent (wt%) to about 60 wt%, with about 35 wt% to about 50 wt% graphite powder preferred. Possible graphite powders include: AIRCO® 60 Graphite Powder produced by Carbide/Graphite Group, Inc., of St. Marys, Pennsylvania; Asbury 4234 Graphite Powder produced by Asbury Graphite Mills, Inc., Asbury, New Jersey; and Dixon 200-42 Graphite Powder produced by Dixon Ticonderoga, Lakehurst, New Jersey; mixtures thereof; and other conventional graphite powders.

The graphite powder is combined with reinforcing fibers which impart structural integrity to the final electrolyte reservoir plate. The ultimate strength of the electrolyte reservoir plate is governed by the amount of bonded reinforcing carbon fiber surface area. If the reinforcing fiber diameter is significantly increased to over about 15μ, less surface area per unit weight of fiber is available for bonding during the thermoset process (discussed below). However, if the reinforcing fiber diameter is significantly below about 5μ, excessive amounts of thermosetting resin are required to bond the electrolyte reservoir plate together. Consequently, these fibers typically have a diameter less than about 15μ, with a fiber diameter of about 5μ to about 10μ preferred.

The reinforcing fiber also preferably has a tensile modulus above about $138 \cdot 10^9$ $N \cdot m^{-2}$ (20 MMpsi). When the tensile modulus of the reinforcing fibers falls below about $138 \cdot 10^9$ $N \cdot m^{-2}$ (20 MMpsi), the flexural strength of the composite is no longer governed by the bonded surface area of the reinforcing fiber but by its tensile strength, and the structural integrity of the electrolyte reservoir plate is reduced to a flexural strength below about 689 $N \cdot cm^{-2}$ (1000 psi). Consequently, fibers possessing a tensile modulus in excess of about $138 \cdot 10^{-9}$ $N \cdot m^{-2}$ (20 MMpsi) can be employed, with fibers possessing a tensile modulus equal to or in excess of about $207 \cdot 10^9$ $N \cdot m^{-2}$ (30 MMpsi) preferred.

In addition to fiber diameter and tensile modulus, the fiber length effects the ability of the reinforcing fibers to impart structural integrity to the electrolyte reservoir plate. Fiber lengths in excess of about 1 mm (0.04 inches) are preferred, with a length of about 2.54 mm (0.10 inches) to about 6.35 mm (0.25 inches)especially preferred. For a horizontal wire papermaking machine, for example, fiber lengths exceeding about 6.35 mm (0.25 inches) are typically undesirable because they diminish the uniformity of the planar sheets due to fiber bundling. Note, longer fibers may not diminish the uniformity of planar sheets formed on other papermaking machines which are more capable of handling long fibers (i.e. inclined wire papermaking machines).

Possible reinforcing fibers include, but are not limited to, carbon fibers such as polyacrylonitrile-based carbon fibers; FORTAFIL® produced by Fortafil Fiber, Inc., of Rockwood, Tennessee, Thornel produced by Amoco Performance Products, Inc., of Ridgefield, Connecticut, RK produced by RK Carbon Fibres Limited of Cheshire, England, AS-4 produced by Hercules Advanced Materials and Systems Co., of Magna Utah, PANEX® produced by Zoltek Corporation of St. Louis, Missouri, mixtures thereof, and other conventional reinforcing fibers which are compatible with the fuel cell environment.

The concentration of reinforcing fibers in the final electrolyte reservoir plate is typically up to about 20 wt%, with up to about 10 wt% preferred, and about 2.5 wt% to about 7.5 wt% especially preferred. Even though the flexural strength of the resultant electrolyte reservoir plate is significantly increased by the addition of the reinforcing fibers, it is feasible to exclude the reinforcing fibers for the purpose of minimizing material costs. Electrolyte reservoir plates made without reinforcing fibers may limit the cell size (i.e. planform) because scrap rates increase substantially as the planform is increased on parts with low flexural strength.

Since the electrolyte reservoir plate is produced by forming planar sheets which are laminated together, cellulosic fibers are also mixed with the graphite powder and reinforcing fibers to provide sufficient wet-strength during the papermaking process such that the planar sheets are strong enough to traverse the entire papermaking machine without breaking. Typically, about 10 wt% to about 30 wt% cellulosic fibers are present in the final electrolyte reservoir plate, with about 15 wt% to about 27 wt% preferred. These fibers are preferably small enough to form a substantially uniform sheet while large enough to provide sufficient hydrogen bonding to impart wet strength during papermaking. Consequently, these fibers have fiber diameters of about 30μ to about 45μ and lengths of about 2 mm to about 4 mm. Cellulosic fibers include fibers derived from natural sources such as hardwoods, softwoods, cotton, and hemp or synthetic materials such as rayon, mixtures thereof, and others, with soft wood pulp preferred. Some such fibers include, but are not limited to Prince George, Northern, Semi-bleached Softwood Pulp, available from Canfor of Vancouver, British Columbia; Brunswick, Southern softwood pulp available from Georgia Pacific of Brunswick, Georgia; Columbus, Southern softwood pulp, available from Weyerhaeuser, of Columbus, Mississippi, mixtures thereof, and other conventional cellulosic fibers.

The mixture of graphite powder, reinforcing fibers, and cellulosic fibers is bonded together with a thermosetting resin which, upon carbonization and graphitization, imparts sufficient structural integrity to the composite to hold the composite together during processing. Typically, thermosetting resins which yield about 40% carbon or greater upon carbonization will impart sufficient structural integrity to the composite post graphitization and provide electrical and thermal continuity between the graphite particles in the electrolyte reservoir plate. Resins with carbon yields below about 40% will likely produce graphitized electrolyte reservoir plate's having marginal strength; below about 689 $N \cdot cm^{-2}$ (1000 psi) flexural strength. There is no known detrimental effect for incorporating higher carbon yield resins. The thermosetting resins include: phenolic resins, polyimides petroleum pitches, and furfuryl alcohols, with phenolic resins preferred. For example, PLENCO™ phenolic resin produced by Plastics Engineering Company, Sheboygan, Wisconsin, and OXYCHEM® phenolic resin produced by Oxychem, Durez Division, North Tonawanda, New York, mixtures thereof, and others. The concentration of thermosetting resin in the final electrolyte reservoir plate typically ranges from about 10 wt% to about 40 wt%, with about 25 wt% to about 30 wt% preferred. This thermosetting resin can be utilized either in the form of a powder preferably having particle sizes below about 20μ, or dispersed in a solvent such as water or an organic solvent such as methanol or ethanol.

Production of the electrolyte reservoir plate comprises forming the solid constituents, the graphite powder, reinforcing fibers, cellulosic fibers, and thermosetting resin, into a slurry using a liquid compatible with the solids. Generally, the liquid is water or a water based liquid. Sufficient liquid to substantially evenly distribute the solid constituents onto the screen of the papermaking machine upon which they are showered, is preferred. Typically, sufficient liquid corresponds to about 90 v/o (volume percent) liquid or greater with about 99 v/o liquid preferred.

The slurry is formed into a planar sheet at approximately 100 lineal feet per minute using a conventional papermaking machine. The slurry is showered substantially evenly onto the horizontal moving screen such that the solid constituents are retained on the screen while the liquid is allowed to pass through. The moving screen travels over a sufficient amount of vacuum sources, or other conventional means, which serve to further dry the retained solid constituents and to promote hydrogen bonding between the cellulosic fibers in the planar sheet. Once the planar sheet is sufficiently dry to support itself, it leaves the screen and travels over several rollers where it is additionally supported by felts running over the rollers. From the rollers the planar sheet travels over a series of heated drums, typically steam or oil heated drums, where residual moisture is volatilized. The dried planar sheet is then spooled onto cardboard tubes for collection. The resultant planar sheet is about 0.508 mm (millimeters; 0.02 inches) to about 1.5 mm (0.06 inches) thick and can be made at any width the particular papermaking machine is capable of forming.

The planar sheet is dried at a temperature sufficient to dry the planar sheet without beginning to cure the thermosetting resin. Typically, the drying temperature ranges from about 93°C (about 200°F) to about 149°C (about 300°F), with about 107°C (about 225°F) to about 135°C (about 275°F) preferred. Once the planar sheet has been dried it is cut to the desired sizes of main sheets **4** and edge strips **6** and laminated. (see Figures 1 and 2) Generally, electrolyte reservoir plates, as with other fuel cell components, are about 89 cm (about 35 inches) by about 89 cm (35 inches) or about 114 cm (about 45 inches) by about 114 cm (about 45 inches).

Lamination comprises laying-up the main sheets **4** on top of one-another with additional edge strips **6** disposed therebetween. The main sheets **4** have four edges, **10a**, **10b**, **10c**, **10d**. The perimeter of the main sheets **4** at two of the opposing edges **10a** and **10c**, are denser areas formed by the edge strips **6** to prevent gas flow through those two edges of the final electrolyte reservoir plate. The width of the edge strips **6** is dependent upon the specific area in which restriction of gas flow is desired, while the length of the edge strips **6** is typically substantially equivalent to the length of the edge of the main sheet where it is located, i.e. the length of **10c**.

The lay-up may consist of virtually any number of main sheets **4** and edge strips **6** wherein the resultant electrolyte reservoir plate has sufficiently densified opposing edges **10a** and **10c** to prevent gas diffusion. The preferred ratio of edge strips to main sheets is about 0.25 to about 0.5. Higher ratios of edge strips result in very dense edges which would be thicker than the remaining areas of the sheets. This non-uniform thickness can cause stacking problems in the fuel cell and migration problems due to voids formed by the non-uniform electrolyte reservoir plates. Additionally, such very dense edges tend to blister during heat treat due to inadequate porosity to remove by-products of the curing process.

The lay-up **2** is laminated by placing it within a molding press and compressing to the desired thickness of about 1.27 mm (about 0.05 inches) to about 3.81 mm (about 0.15 inches) under an axial load of up to about 2077 $\frac{N}{cm^2}$ (3,000 psig) and a temperature of about 150°C (300°F) to about 230°C (450°F), with a temperature of about 165°C (325°F) to about 190°C (375°F) preferred for about 1 to about 15 minutes. The laminated lay-up is then carbonized by heating at about 8°F/hour to about 815°C (1,500°F) to about 1095°C (2,000°F) and remaining at that temperature for about 0.5 hours to about 4 hours, and subsequently graphitized at about 2000°C (3,632°F) to about 3000°C (5,432°F) for about 2 hours to about 4 hours.

It should be noted that the planar sheets can be formed from the graphite powder, reinforcing fibers, and cellulosic fibers. In such a case, once the planar sheets have been formed and dried, they can be impregnated with the thermosetting resin.

The invention will be further clarified with reference to the following illustrative examples. These examples are meant to illustrate the process of forming the laminated electrolyte reservoir plate of the present invention.

Example I

The following process can be utilized to form a 40 wt% AirCo 60 graphite powder, 5 wt% FORTAFIL 3.175 mm (⅛ inch) unsized carbon fiber, 28 wt% OXYCHEM Phenolic resin, and 27 wt% Softwood Pulp.

1. Water is mixed with the solids in a portion of 0.4 g graphite powder, 0.05 g carbon fibers, 0.28 g OxyChem Phenolic Resin, and 0.27 g Softwood Pulp to form a slurry having about 1 v/o solids.
2. Once thoroughly blended, the slurry is showered onto a horizontally moving screen to form a planar sheet with a basis weight of 113.4 kg/ream (250 lb /ream) of 405.6 g·m^{-2} (12 oz/sq.yard).
3. The screen is passed over a vacuum to remove some of the remaining water, and thereby dry the planar sheet.
4. The dried planar sheet is then directed over rollers and oil heated drums to volatilize residual water and form the dried paper. The drums are heated to 121°C (250°F).
5. The dried paper is spooled on a cardboard tube for collection.
6. The spooled paper is cut into 254 mm - 889 mm (10 - 35 inch) by 889 mm (35 inch) sheets and 152.4 mm - 889 mm (6 - 35 inch) by 71.9 mm (2.83 inch) strips.
7. The sheets and strips are then laid up such that strips are laid on opposed edges of the sheets between sheets 4 and 5, 5 and 6, and 6 and 7.
8. The lay-up is then compression molded to 3.56 mm (0.140 inches) at 1643 N·cm^{-2} (2370 psig) for 5 minutes at about 175°C (345°F) to laminate the lay-up.
9. The laminated lay-up is carbonized in a nitrogen environment up to about 820°C (1510°F) at 4.44°C (8°F) per hour and subsequently graphitized at 2350°C at 42.8°C/hour (4262°F at 77°F/hour).

Example II

The following process can be employed to form an electrolyte reservoir plate having 40 wt% AirCo 60 graphite powder, 5 wt% Fortafil 3.175 mm (⅛ inch) unsized carbon fiber, 28 wt% OxyChem Phenolic resin, and 27 wt% Softwood

Pulp.

1. A slurry is prepared and formed into paper as described in Example I steps 1-5.
2. The paper is cut into 254 mm - 1.143 m (10 - 45 inch) by 1.143 m (45 inch) sheets 304.8 mm - 1.143 m (12 - 45 inch) by 71.9 mm (2.83 inch) strips.
3. The sheets and strips are then laid up such that 2 strips are laid on opposed edges of the sheets between sheets 4 and 5, 5 and 6, and 6 and 7.
4. The lay-up is laminated in a compression mold to about 3.56 mm (0.140 inches) at 995.4 N·cm^{-2} (1430 psig) for 5 minutes at about 170°C (340°F).
5. The laminated lay-up is carbonized in a nitrogen environment at about 820°C (1510°F) at 4.44°C (8°F) per hour and subsequently graphitized at 2350°C at 42.8°C/hour (4262°F at 77°F/hour).

There are numerous advantages realized by the electrolyte reservoir plate of the present invention. Due to the dense edge seals which are about 25% to about 50% more dense than the remainder of the electrolyte reservoir plate, impregnation of the edge seals with hydrophilic ink or similar material is not required. In addition to improved edge seals, the electrolyte reservoir plate has improved structural integrity due to the use of longer reinforcing fibers than prior art electrolyte reservoir plates. These improvements over the prior art are illustrated in the following Table which compares the prior art electrolyte reservoir plate (ERP) to the laminated ERP of the present invention.

Physical Properties (Post Graphitization)	Prior Art ERP	Laminated ERP
IR (mV/mil at 100 ASF)	0.004	0.011
IR (mV/µm at 9.29 A/m^2)	0.1016	0.2794
Flexural Strength (psi)	844	2461
Flexural Strength (N/cm^2)	581.5	1695.6
Flexural Modulus (psi x 10^{-6})	0.254	0.67
Flexural Modulus (N/cm^2)	0.1750	0.4616
Compressive Strength (psi)	221	317
Compressive Strength (N/cm^2)	152.3	218.4
Compressive Modulus (psi)	12356	9026
Compressive Modulus (N/cm^2)	8513.3	6218.9
Mean Pore Size (µ)	18.2	13.6
Porosity (%)	43.0	45.0

Claims

1. A laminated electrolyte reservoir plate, comprising: a mixture of graphite powder, reinforcing fibers, cellulosic fibers, and thermosetting resin formed into a planar sheet, wherein said sheet has been cut into a plurality of main sheets having at least two opposing edges and a plurality of edge strips which have been laid-up such that said edge strips are located along the perimeter of said main sheet along said opposing edges, laminated together, carbonized, and graphitized.

2. A laminated electrolyte reservoir plate as in Claim 1, wherein said reinforcing fibers are carbon fibers.

3. A laminated electrolyte reservoir plate as in Claim 1, wherein said cellulosic fibers are derived from natural materials, synthetic materials, or mixtures thereof.

4. A laminated electrolyte reservoir plate as in Claim 3, wherein said natural materials are hardwoods, softwoods, cotton, hemp, or mixtures thereof, and said synthetic material is rayon.

5. A laminated electrolyte reservoir plate as in Claim 1, wherein said thermosetting resin is a phenolic resin, polyimide, petroleum pitch, furfuryl alcohol, or mixture thereof.

6. A laminated electrolyte reservoir plate as in Claim 1 comprising about 25 wt% to about 60 wt% graphite powder.

7. A laminated electrolyte reservoir plate as in Claim 1 comprising up to about 20 wt% reinforcing fibers.

8. A laminated electrolyte reservoir plate as in Claim 1 comprising about 10 wt% to about 30 wt% cellulosic fibers.

9. A laminated electrolyte reservoir plate as in Claim 1 comprising about 10 wt% to about 40 wt% thermosetting resin.

10. A method for forming a laminated electrolyte reservoir plate, comprising the steps of:

a. mixing graphite powder, reinforcing fibers, cellulosic fibers, and thermosetting resin with a liquid to form a slurry;
b. forming said slurry into a planar sheet;
c. removing said liquid from said planar sheet;
d. cutting said planar sheet into a plurality of main sheets having at least two opposing edges and a plurality of edge strips;
e. laying-up said main sheets and said edge strips such that said edge strips are located along the perimeter of said main sheets adjacent to said opposing edges; and
f. laminating, carbonizing, and graphitizing said lay-up to form the laminated electrolyte reservoir plate.

11. A method for forming a laminated electrolyte reservoir plate as in Claim 10 wherein said slurry is about 90 v/o liquid or greater.

12. A method for forming a laminated electrolyte reservoir plate as in Claim 10 wherein said slurry is formed into a planar sheet by showering said slurry onto a screen.

13. A method for forming a laminated electrolyte reservoir plate as in Claim 10 wherein said liquid is removed from said planar sheet with at least one vacuum and/or with heat.

14. A method for forming a laminated electrolyte reservoir plate as in Claim 10 wherein said lay-up is laminated at about 150°C to about 230°C.

15. A method for forming a laminated electrolyte reservoir plate as in Claim 10 wherein said lay-up is laminated under an axial load of up to about 2077 $N \cdot cm^{-2}$ (3,000 psig).

16. A method for forming a laminated electrolyte reservoir plate as in Claim 10 wherein said lay-up is carbonized at about 815°C to about 1,095°C.

17. A method for forming a laminated electrolyte reservoir plate as in Claim 10 wherein said lay-up is graphitized at about 2,000°C to about 3,000°C.

18. A method for forming a laminated electrolyte reservoir plate as in Claim 10 wherein said lay-up is laminated under an axial load of up to about 2077 $N \cdot cm^{-2}$ (3,000 psig), a temperature of about 150°C to about 190°C for about 1 minute to about 15 minutes.

19. A method for forming a laminated electrolyte reservoir plate as in Claim 10 wherein said main sheets have opposed parallel edges and said edge strips are laid-up such that said edge strips are disposed between said main sheets adjacent to said parallel edges.

20. A method for forming a laminated electrolyte reservoir plate, comprising the steps of:

a. mixing graphite powder, reinforcing fibers, and cellulosic fibers, with a liquid to form a slurry;
b. forming said slurry into a planar sheet;
c. removing said liquid from said planar sheet to form a dried sheet;
d. impregnating said dried sheet with thermosetting resin;

e. cutting said planar sheet into a plurality of main sheets having at least two opposing edges and a plurality of edge strips;

f. laying-up said main sheets and said edge strips such that said edge strips are located along the perimeter of said main sheets adjacent to said opposing edges; and

g. laminating, carbonizing, and graphitizing said lay-up to form the laminated electrolyte reservoir plate.

21. A method for forming a laminated electrolyte reservoir plate as in Claim 20 wherein said slurry is about 90 v/o liquid or greater.

22. A method for forming a laminated electrolyte reservoir plate as in Claim 20 wherein said slurry is formed into a planar sheet by showering said slurry onto a screen.

23. A method for forming a laminated electrolyte reservoir plate as in Claim 20 wherein said liquid is removed from said planar sheet with at least one vacuum and/or with heat.

24. A method for forming a laminated electrolyte reservoir plate as in Claim 20 wherein said lay-up is laminated at about 150°C to about 230°C.

25. A method for forming a laminated electrolyte reservoir plate as in Claim 20 wherein said lay-up is laminated under an axial load of up to about 2077 $N \cdot cm^{-2}$ (3,000 psig).

26. A method for forming a laminated electrolyte reservoir plate as in Claim 20 wherein said lay-up is carbonized at about 815°C to about 1,095°C.

27. A method for forming a laminated electrolyte reservoir plate as in Claim 20 wherein said lay-up is graphitized at about 2,000°C to about 3,000°C.

28. A method for forming a laminated electrolyte reservoir plate as in Claim 20 wherein said lay-up is laminated under an axial load of up to about 2077 $N \cdot cm^{-2}$ (3,000 psig), a temperature of about 150°C to about 190°C for about 1 minute to about 15 minutes.

29. A method for forming a laminated electrolyte reservoir plate as in Claim 20 wherein said main sheets have opposed parallel edges and said edge strips are laid-up such that said edge strips are disposed between said main sheets adjacent to said parallel edges.

Patentansprüche

1. Elektrolytreservoir-Schichtplatte aufweisend: ein zu einem ebenen Flächengebilde geformtes Gemisch aus Graphitpulver, Verstärkungsfasern, Cellulosefasern und wärmehärtbarem Harz, wobei das Flächengebilde in eine Mehrzahl von Haupt-Flächengebilden mit mindestens zwei entgegengesetzten Rändern und eine Mehrzahl von Randstreifen geschnitten wurde, die dergestalt aufeinandergelegt wurden, daß die Randstreifen längs der einander entgegengesetzten Ränder am Umfang des Haupt-Flächengebildes angeordnet sind, zusammenlaminiert, carbonisiert und graphitisiert wurden.

2. Elektrolytreservoir-Schichtplatte nach Anspruch 1, bei der die Verstärkungsfasern Kohlenstoffasern sind.

3. Elektrolytreservoir-Schichtplatte nach Anspruch 1, bei der die Cellulosefasern von natürlichen Materialien, synthetischen Materialien oder Gemischen davon abstammen.

4. Elektrolytreservoir-Schichtplatte nach Anspruch 3, bei der die natürlichen Materialien Harthölzer, Weichhölzer, Baumwolle, Hanf oder Gemische davon sind, und das synthetische Material Reyon ist.

5. Elektrolytreservoir-Schichtplatte nach Anspruch 1, bei der das wärmehärtbare Harz ein Phenolharz, ein Polyimid, Petrolpech, Furfürylalkohol oder ein Gemisch davon ist.

6. Elektrolytreservoir-Schichtplatte nach Anspruch 1, etwa 25 Gew% bis etwa 60 Gew% Graphitpulver aufweisend.

7. Elektrolytreservoir-Schichtplatte nach Anspruch 1, aufweisend bis zu etwa 20 Gew% Verstärkungsfasern.

8. Elektrolytreservoir-Schichtplatte nach Anspruch 1, aufweisend etwa 10 Gew% bis etwa 30 Gew% Cellulosefasern.

9. Elektrolytreservoir-Schichtplatte nach Anspruch 1, aufweisend etwa 10 Gew% bis etwa 40 Gew% wärmehärtbares Harz.

10. Verfahren zur Herstellung einer Elektrolyreservoir-Schichtplatte, folgende Schritte aufweisend:

a. Mischen von Graphitpulver, Verstärkungsfasern, Cellulosefasern und wärmehärtbarem Harz mit einer Flüssigkeit zur Bildung einer Aufschlämmung;
b. Formen der Aufschlämmung zu einem ebenen Flächengebilde;
c. Entfernen der Flüssigkeit aus dem ebenen Flächengebilde;
d. Schneiden des ebenen Flächengebildes in eine Mehrzahl von Haupt-Flächengebilden mit mindestens zwei entgegengesetzten Rändern und eine Mehrzahl von Randstreifen;
e. Aufeinanderlegen der Haupt-Flächengebilde und der Randstreifen dergestalt, daß die Randstreifen am Umfang der Haupt-Flächengebilde nahe an den einander entgegengesetzten Rändern angeordnet werden; und
f. Laminieren, Carbonisieren und Graphitisieren der Schichtung zur Herstellung der Elektrolytreservoir-Schichtplatte.

11. Verfahren zur Herstellung einer Elektrolytreservoir-Schichtplatte nach Anspruch 10, bei dem die Aufschlämmung zu etwa 90 Vol% oder mehr Flüssigkeit ist.

12. Verfahren zur Herstellung einer Elektrolytreservoir-Schichtplatte nach Anspruch 10, bei dem die Aufschlämmung durch Rieseln der Aufschlämmung auf ein Sieb zu einem ebenen Flächengebilde geformt wird.

13. Verfahren zur Herstellung einer Elektrolytreservoir-Schichtplatte nach Anspruch 10, bei dem die Flüssigkeit mittels mindestens eines Vakuums und/oder mittels Wärme aus dem ebenen Flächengebilde entfernt wird.

14. Verfahren zur Herstellung einer Elektrolytreservoir-Schichtplatte nach Anspruch 10, bei dem die Schichtung bei etwa 150°C bis etwa 230°C laminiert wird.

15. Verfahren zur Herstellung einer Elektrolytreservoir-Schichtplatte nach Anspruch 10, bei dem die Schichtung unter einer Axiallast von bis zu etwa 2077 N • cm^{-2} (3000 psig) laminiert wird.

16. Verfahren zur Herstellung einer Elektrolytreservoir-Schichtplatte nach Anspruch 10, bei dem die Schichtung bei etwa 815°C bis etwa 1095°C carbonisiert wird.

17. Verfahren zur Herstellung einer Elektrolytreservoir-Schichtplatte nach Anspruch 10, bei dem die Schichtung bei etwa 2000°C bis etwa 3000°C graphitisiert wird.

18. Verfahren zur Herstellung einer Elektrolytreservoir-Schichtplatte nach Anspruch 10, bei dem die Schichtung unter einer Axiallast von bis zu etwa 2077 N • cm^{-2} (3000 psig), einer Temperatur von etwa 150°C bis etwa 190°C etwa 1 Minute bis etwa 15 Minuten lang laminiert wird.

19. Verfahren zur Herstellung einer Elektrolytreservoir-Schichtplatte nach Anspruch 10, bei dem die Haupt-Flächengebilde einander entgegengesetzte parallele Ränder haben und die Randstreifen dergestalt aufeinandergelegt werden, daß die Randstreifen zwischen den Haupt-Flächengebilden nahe an den parallelen Rändern angeordnet werden.

20. Verfahren zur Herstellung einer Elektrolytreservoir-Schichtplatte, folgende Schritte aufweisend:

a. Mischen von Graphitpulver, Verstärkungsfasern und Cellulosefasern mit einer Flüssigkeit zur Bildung einer Aufschlämmung;
b. Formen der Aufschlämmung zu einem ebenen Flächengebilde;
c. Entfernen der Flüssigkeit aus dem ebenen Flächengebilde zur Herstellung eines getrockneten Flächengebildes;

d. Imprägnieren des getrockneten Flächengebildes mit wärmehärtbarem Harz;
e. Schneiden des ebenen Flächengebildes in eine Mehrzahl von Haupt-Flächengebilden mit mindestens zwei einander entgegengesetzten Rändern und eine Mehrzahl von Randstreifen;
f. Aufeinanderlegen der Haupt-Flächengebilde und der Randstreifen dergestalt, daß die Randstreifen am Umfang der Haupt-Flächengebilde nahe an den einander entgegengesetzten Rändern angeordnet werden; und
g. Laminieren, Carbonisieren und Graphitisieren der Schichtung zur Herstellung der Elektrolytreservoir-Schichtplatte.

21. Verfahren zur Herstellung einer Elektrolytreservoir-Schichtplatte nach Anspruch 20, bei dem die Aufschlämmung zu etwa 90 Vol% oder mehr Flüssigkeit ist.

22. Verfahren zur Herstellung einer Elektrolytreservoir-Schichtplatte nach Anspruch 20, bei dem die Aufschlämmung durch Rieseln der Aufschlämmung auf ein Sieb zu einem ebenen Flächengebilde geformt wird.

23. Verfahren zur Herstellung einer Elektrolytreservoir-Schichtplatte nach Anspruch 20, bei dem die Flüssigkeit mittels mindestens eines Vakuums und/oder mittels Wärme aus dem ebenen Flächengebilde entfernt wird.

24. Verfahren zur Herstellung einer Elektrolytreservoir-Schichtplatte nach Anspruch 20, bei dem die Schichtung bei etwa 150°C bis etwa 230°C laminiert wird.

25. Verfahren zur Herstellung einer Elektrolytreservoir-Schichtplatte nach Anspruch 20, bei dem die Schichtung unter einer Axiallast von bis zu etwa 2077 N • cm^{-2} (3000 psig) laminiert wird.

26. Verfahren zur Herstellung einer Elektrolytreservoir-Schichtplatte nach Anspruch 20, bei dem die Schichtung bei etwa 815°C bis etwa 1095°C carbonisiert wird.

27. Verfahren zur Herstellung einer Elektrolyreservoir-Schichtplatte nach Anspruch 20, bei dem die Schichtung bei etwa 2000°C bis etwa 3000°C graphitisiert wird.

28. Verfahren zur Herstellung einer Elektrolytreservoir-Schichtplatte nach Anspruch 20, bei dem die Schichtung unter einer Axiallast von bis zu etwa 2077 N • cm^{-2} (3000 psig), einer Temperatur von etwa 150°C bis etwa 190°C etwa 1 Minute bis etwa 15 Minuten lang laminiert wird.

29. Verfahren zur Herstellung einer Elektrolytreservoir-Schichtplatte nach Anspruch 20, bei dem die Haupt-Flächengebilde einander entgegengesetzte parallele Ränder haben und die Randstreifen dergestalt aufeinandergelegt werden, daß die Randstreifen zwischen den Haupt-Flächengebilden nahe an den parallelen Rändern angeordnet werden.

Revendications

1. Plaque stratifiée de réservoir d'électrolyte, comprenant : un mélange de poudre de graphite, des fibres de renforcement, des fibres cellulosiques, et de la résine thermodurcissable façonnée en une feuille plane, dans laquelle ladite feuille a été coupée en une pluralité de feuilles principales ayant au moins deux bords opposés et une pluralité de bandes de bordure qui ont été superposées de telle façon que lesdites bandes de bordure soient situées le long du périmètre de ladite feuille principale le long de lesdits bords opposés, stratifiées ensemble, carbonisées, et graphitisées.

2. Plaque stratifiée de réservoir d'électrolyte selon la revendication 1, dans laquelle lesdites fibres de renforcement sont des fibres de carbone.

3. Plaque stratifiée de réservoir d'électrolyte selon la revendication 1, dans laquelle lesdites fibres cellulosiques sont dérivées de matériaux naturels, de matériaux synthétiques, ou de mélanges de ceux-ci.

4. Plaque stratifiée de réservoir d'électrolyte selon la revendication 3, dans laquelle lesdits matériaux naturels sont des bois durs, des bois tendres, du coton, du chanvre, ou des mélanges de ceux-ci, et ledit matériau synthétique est de la rayonne.

5. Plaque stratifiée de réservoir d'électrolyte selon la revendication 1, dans laquelle ladite résine thermodurcissable est une résine phénolique, un polyimide, du brai de pétrole, de l'alcool furfurylique, ou un mélange de ceux-ci.

6. Plaque stratifiée de réservoir d'électrolyte selon la revendication 1 comprenant environ 25 % en poids à environ 60 % en poids de poudre de graphite.

7. Plaque stratifiée de réservoir d'électrolyte selon la revendication 1 comprenant jusqu'à environ 20 % en poids des fibres de renforcement.

8. Plaque stratifiée de réservoir d'électrolyte selon la revendication 1 comprenant environ 10 % en poids à environ 30 % en poids de fibres cellulosiques.

9. Plaque stratifiée de réservoir d'électrolyte selon la revendication 1 comprenant environ 10 % en poids à environ 40 % en poids de résine thermodurcissable.

10. Procédé pour former une plaque stratifiée de réservoir d'électrolyte, comprenant les opérations consistant à:

a. mélanger de la poudre de graphite, des fibres de renforcement, des fibres cellulosiques, et de la résine thermodurcissable avec un liquide pour former une boue;
b. façonner ladite boue en une feuille plane;
c. enlever ledit liquide de ladite feuille plane;
d. couper ladite feuille plane en une pluralité de feuilles principales ayant au moins deux bords opposés et une pluralité de bandes de bordure;
e. superposer lesdites feuilles principales et lesdites bandes de bordure de telle façon que lesdites bandes de bordure soient situées le long du périmètre desdites feuilles principales adjacentes auxdits bords opposés; et
f. stratifier, carboniser, et graphitiser ledit empilement pour former la plaque stratifiée de réservoir d'électrolyte.

11. Procédé pour former une plaque stratifiée de réservoir d'électrolyte selon la revendication 10 dans laquelle ladite boue est liquide à environ 90 % en volume ou plus.

12. Procédé pour former une plaque stratifiée de réservoir d'électrolyte selon la revendication 10 dans laquelle ladite boue est façonnée en une feuille plane en répandant ladite boue sur un tamis.

13. Procédé pour former une plaque stratifiée de réservoir d'électrolyte selon la revendication 10 dans laquelle ledit liquide est enlevé de ladite feuille plane avec au moins une aspiration et/ou avec de la chaleur.

14. Procédé pour former une plaque stratifiée de réservoir d'électrolyte selon la revendication 10 dans laquelle ledit empilement est stratifié à environ 150 °C jusqu'à environ 230°C.

15. Procédé pour former une plaque stratifiée de réservoir d'électrolyte selon la revendication 10 dans laquelle ledit empilement est stratifié sous une charge axiale allant jusqu'à environ 2.077 $N{\cdot}cm^{-2}$ (3,000 psig).

16. Procédé pour former une plaque stratifiée de réservoir d'électrolyte selon la revendication 10 dans laquelle ledit empilement est carbonisé à environ 815 °C jusqu'à environ 1.095°C.

17. Procédé pour former une plaque stratifiée de réservoir d'électrolyte selon la revendication 10 dans laquelle ledit empilement est graphitisé à environ 2.000 °C jusqu'à environ 3.000 °C.

18. Procédé pour former une plaque stratifiée de réservoir d'électrolyte selon la revendication 10 dans laquelle ledit empilement est stratifié sous une charge axiale allant jusqu'à environ 2.077 $N{\cdot}cm^{-2}$ (3,000 psig), à une température d'environ 150 °C à environ 190 °C pendant environ 1 minute à environ 15 minutes.

19. Procédé pour former une plaque stratifiée de réservoir d'électrolyte selon la revendication 10 dans laquelle lesdites feuilles principales ont des bords opposées parallèles et des bandes de bordure sont superposées de telle façon que lesdites bandes de bordure soient disposées entre lesdites feuilles principales adjacentes auxdits bords parallèles.

20. Procédé pour former une plaque stratifiée de réservoir d'électrolyte, comprenant les opérations consistant à :

a. mélanger de la poudre de graphite, des fibres de renforcement, et des fibres cellulosiques, avec un liquide pour former une boue ;
b. façonner ladite boue en une feuille plane;
c. enlever ledit liquide de ladite feuille plane pour former une feuille séchée;
d. imprégner ladite feuille sèche de résine thermodurcissable;
e. couper ladite feuille plane en une pluralité de feuilles principales ayant au moins deux bords opposés et une pluralité de bandes de bordure;
f. superposer lesdites feuilles principales et lesdites bandes de bordure de telle façon que lesdites bandes de bordure soient situées le long du périmètre desdites feuilles principales adjacentes auxdits bords opposés; et
g. stratifier, carboniser, et graphitiser ledit empilement pour former la plaque stratifiée de réservoir d'électrolyte.

21. Procédé pour former une plaque stratifiée de réservoir d'électrolyte selon la revendication 20 dans laquelle ladite boue est liquide à environ 90 % en volume ou plus.

22. Procédé pour former une plaque stratifiée de réservoir d'électrolyte selon la revendication 20 dans laquelle ladite boue est façonnée en une feuille plane en répandant ladite boue sur un tamis.

23. Procédé pour former une plaque stratifiée de réservoir d'électrolyte selon la revendication 20 dans laquelle ledit liquide est enlevé de ladite feuille plane avec au moins une aspiration et/ou avec de la chaleur.

24. Procédé pour former une plaque stratifiée de réservoir d'électrolyte selon la revendication 20 dans laquelle ledit empilement est stratifié à environ 150 °C jusqu'à environ 230°C.

25. Procédé pour former une plaque stratifiée de réservoir d'électrolyte selon la revendication 20 dans laquelle ledit empilement est stratifié sous une charge axiale allant jusqu'à environ 2.077 $N{\cdot}cm^{-2}$ (3,000 psig).

26. Procédé pour former une plaque stratifiée de réservoir d'électrolyte selon la revendication 20 dans laquelle ledit empilement est carbonisé à environ 815 °C jusqu'à environ 1.095°C.

27. Procédé pour former une plaque stratifiée de réservoir d'électrolyte selon la revendication 20 dans laquelle ledit empilement est graphitisé à environ 2.000 °C jusqu'à environ 3.000 °C.

28. Procédé pour former une plaque stratifiée de réservoir d'électrolyte selon la revendication 20 dans laquelle ledit empilement est stratifié sous une charge axiale allant jusqu'à environ 2.077 $N{\cdot}cm^{-2}$ (3,000 psig), à une température d'environ 150 °C jusqu'à environ 190 °C pendant environ 1 minute à environ 15 minutes.

29. Procédé pour former une plaque stratifiée de réservoir d'électrolyte selon la revendication 20 dans laquelle lesdites feuilles principales ont des bords opposés parallèles et lesdites bandes de bordure sont superposées de telle façon que lesdites bandes de bordure soient disposées entre lesdites feuilles principales adjacentes auxdits bords parallèles.

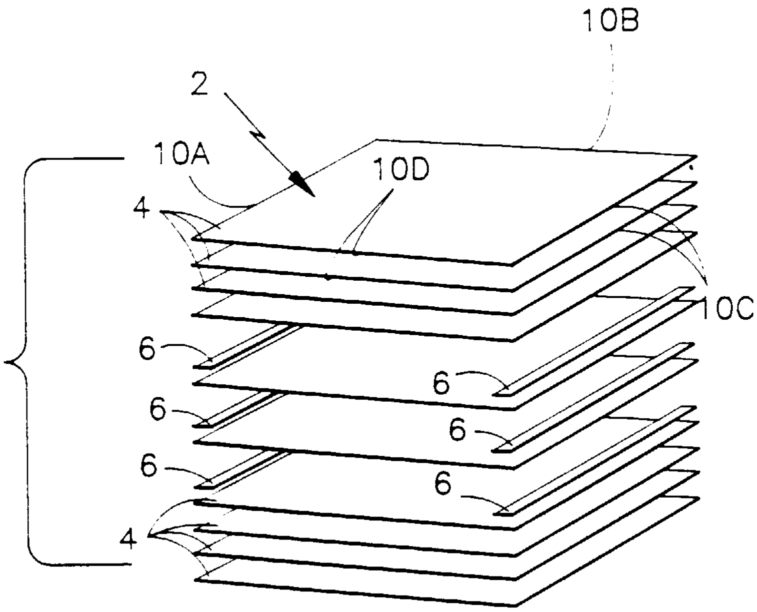

fig. 1

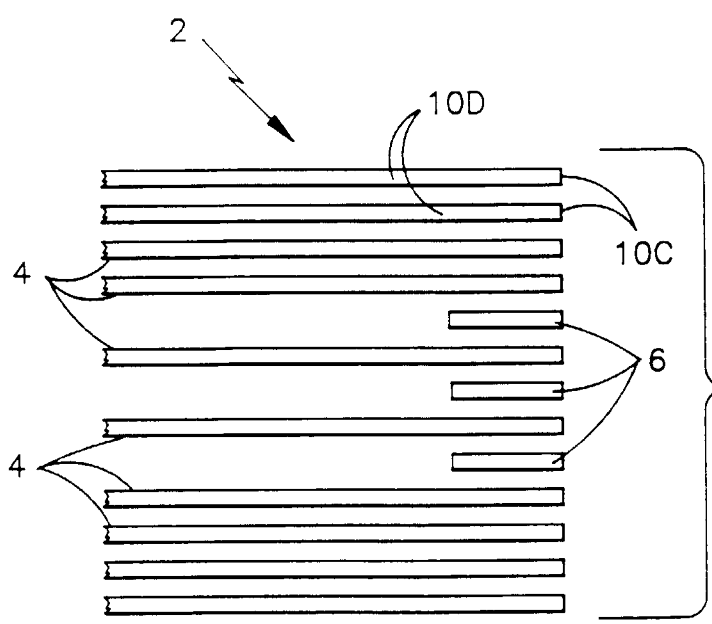

fig. 2